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Electron magnetic chiral dichroism in CrO_2 thin films using monochromatic probe illumination in a transmission electron microscope

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ABSTRACT

Electron magnetic chiral dichroism (EMCD) has been studied in CrO_2 thin films (with (100) and (110) growth orientations on TiO_2 substrates) using a gun monochromator in an aberration corrected transmission electron microscope operating at 300 kV. Excellent signal-to-noise ratio is obtained at spatial resolution ~10 nm using a monochromatic probe as compared to conventional parallel illumination, large area convergent beam electron diffraction and scanning transmission electron microscopy techniques of EMCD. Relatively rapid exposure using mono probe illumination enables collection of EMCD spectra in total of 8–9 min in energy filtered imaging mode for a given Cr $L_{2,3}$ energy scan (energy range ~35 eV). We compared the EMCD signal obtained by extracting the Cr $L_{2,3}$ spectra under three beam diffraction geometry of two different reciprocal vectors (namely g=110 and 200) and found that the g=200 vector enables acquisition of excellent EMCD signal from relatively thicker specimen area due to the associated larger extinction distance. Orbital to spin moment ratio has been calculated using EMCD sum rules for 3*d* elements and dichroic spectral features associated with CrO_2 are compared and discussed with XMCD theoretical spectra.

1. Introduction

Energy-loss electron magnetic chiral dichroism (EMCD) in a transmission electron microscope (TEM) is a new technique to study the magnetism in materials at atomic scale resolution [1]. EMCD is the electron counterpart of the well known x-ray magnetic circular dichroism (XMCD) technique which has been extensively used for studying the element specific orbital and spin magnetic moments of various materials [2–5]. EMCD has the advantage of superior spatial resolution (~2 nm has already been demonstrated) [6,7] and provides bulk information as compared to XMCD which is mostly surface sensitive and difficult to routinely achieve spatial resolution below 100 nm [8]. Recent progress in x-ray optics and soft x-ray magnetic microscopy could improve the lateral resolution to about 15 nm [9]. On the other hand, there is a theoretical prediction of achieving atomic scale resolution EMCD and the possibility to study the magnetic properties of individual atom in the material [10,11], which will be extremely useful in many emerging areas of spintronics. This may be possible with the availability of ultra high resolution aberration corrected TEM for example in a FEI TITAN^{3TM} 80–300 kV microscope. Various experimental methods to perform EMCD have already been demonstrated [12].

In the present report we have performed EMCD experiment by exciting the gun monochromator in a FEI TITAN^{3TM} 80–300 kV transmission electron microscope where high signal to noise ratio with spatial resolution ~10 nm and superior energy resolution ~0.2–0.3 eV (depending on GIF entrance aperture and camera length) can be achieved. This method allows working with the parallel illumination with quicker acquisition time (total ~8–9 min) in energy filtered imaging mode. One can also achieve very high spatial resolution by controlling the condenser slit width (down to 5 nm) under parallel illumination condition. Parallel beam illumination is essential (without range of k vectors) to obtain two or three beam diffraction geometry for simultaneous momentum transfer by direct and diffracted waves (phase shifted by $\pi/2$ between themselves), which after interference gives rise to chiral signal in the presence of a magnetic field in the material. This new method allows achieving near atomic resolution EMCD with improved signals over background in energy spectroscopy imaging (ESI) mode.

We have studied EMCD spectra corresponding to Cr $L_{2,3}$ absorption edge of high quality CrO_2 thin films grown by chemical vapor deposition (CVD) with two different growth orientations, (100) and (110) on TiO_2 substrates [13]. In addition, we have

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studied the effect of reciprocal lattice vectors of two different magnitudes on the EMCD signal for a given TEM sample thickness. CrO_2 is the only known half metal with essentially complete spin polarization at least at low temperatures [14,15]. CrO_2 has a rutile structure (space group D_{4h}^{14}: $P4_2/mnm$) and has inversion symmetry about the body centered Cr atom, which means dichroic signal obtained through mixed dynamic form factor (MDFF) will be due to the presence of magnetic moments alone and not due to lack of inversion symmetry [1]. XMCD studies from similar films revealed higher spin moment for (110) orientation (1.8 μ_B/Cr) as compared to (100) oriented film (1.2 μ_B/Cr) [16]. However, we obtained higher EMCD signal for (100) oriented film as compared to (110) film and this result is in discrepancy with XMCD and SQUID data [16]. The EMCD spectral features of CrO_2 are discussed with respect to the reported XMCD CrO_2 spectra and compared with theoretical calculation based on density functional theory as implemented in WIEN2k code [17].

2. Experimental details

2.1. Gun monochromator parallel illumination condition

Gun monochromator illumination was exploited in EMCD experimentation in order to achieve improved signal and energy resolution. Gun monochromator is based on the principle of applying a static potential that disperses the electron beam depending on its energy spread in a plane perpendicular to its propagation. After dispersion central bright region of the probe has an energy resolution ~0.18 eV at 300 kV [obtained at high magnification (~115 ×), smallest GIF entrance aperture (1 mm) and highest dispersion (0.1 eV/channel) in the spectrometer]. Such a probe has parallel illumination and a small bright central region of the probe ~5–20 nm diameter, can be selected using parallel slit at the height of C1 condenser lens available in FEI TITAN microscope. As the probe is not defocused (spread) to fill the C2 aperture (in TITAN there are three condenser lens system, these are C1, C2 and C3), it therefore contains comparatively large electron flux, which improves signal over noise and thus allows quicker acquisition of spectra.

Fig. 1(a) shows the mono-chromatic probe at the center of the C1 aperture height and Fig. 1(b) shows the size of illumination as selected by the slit perpendicular to the energy spread of the mono probe. EMCD technique demands at least two spots in the diffraction pattern (two beam condition), i.e. one direct beam and another diffracted beam (using crystal as an interferometer) so that simultaneous momentum transfer is possible in order to obtain chiral signal according to the principle first described and demonstrated by Hébert and Schattschneider [18]. The CrO_2 (110) oriented film is about 45 nm thick, as is seen through the slit in the Fig. 1(b). Fig. 1(c) shows the diffraction pattern from the film along (001) zone axis and 200 and 110 spots are indexed. The EMCD signal is extracted in the 3-beam condition by selecting 200 and 110 reciprocal lattice vectors from the positions on the Thales circle as shown in Fig. 2. Cr $L_{2,3}$ spectra between parallel mode illumination (mono focus spread at C1 is ~−24) and mono probe illumination (mono focus spread at C1 is ~0, 0 means converged) with slit are compared in Fig. 3. An improvement ~200,000 (~60%) in terms of counts (intensity) can easily be obtained with mono probe illumination for the same exposure time and area of acquisition. Parallel illumination is important as we now know from previously published results that the EMCD signal degrades with increasing convergence angle (range of k vectors increases with the increase in probe convergence angle) [6] as is the case in a LACDIFF or converged STEM probe acquisition mode. Therefore, small probe (down to few Å) along with parallel illumination (or very little convergence) are required for near atomic resolution (few nm) and better dichroic signal.

Parallel illumination is essential for energy loss diffraction pattern imaging/energy spectroscopic imaging (ESI) where collection angle is very small (of the order of the Bragg angle in a TEM) and condition requiring same convergence and collection angles as small as this cannot be achieved for STEM or LACDIFF mode of EMCD data acquisition. STEM probe (~1.7 nm) has been used to demonstrate ~2 nm spatial resolutions in EMCD experiment [6,7]. This approach requires acquisition of spectra in spectroscopy mode (in the first case) of EELS by physically placing objective aperture at the diffraction plane at two different positions. This technique lacks precise positioning of aperture (small variation of size of the aperture is also not possible with the available apertures in TEMs) and acquisition at two different point of time often result in dissimilar signal to noise ratio which often may not be possible to normalize in order to extract useful

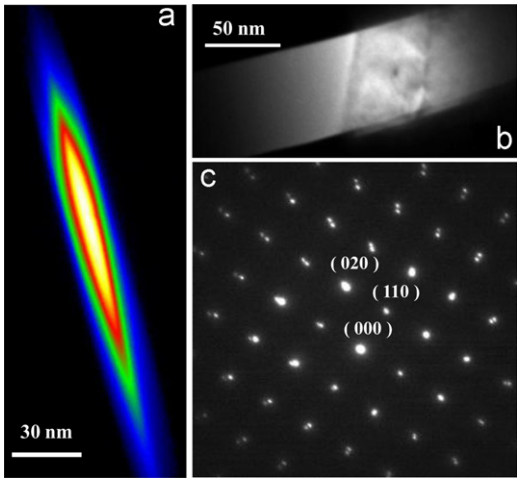

Fig. 1. (a) Mono chromatic probe at the height of C1 lens, (b) Central part of the probe as selected by parallel slit and (c) Selected area diffraction pattern from CrO_2/TiO_2 along 001 zone axis and $<200>$ and $<110>$ spots are indexed. There are also spots appearing from TiO_2 substrate and this shows that the film is relaxed.

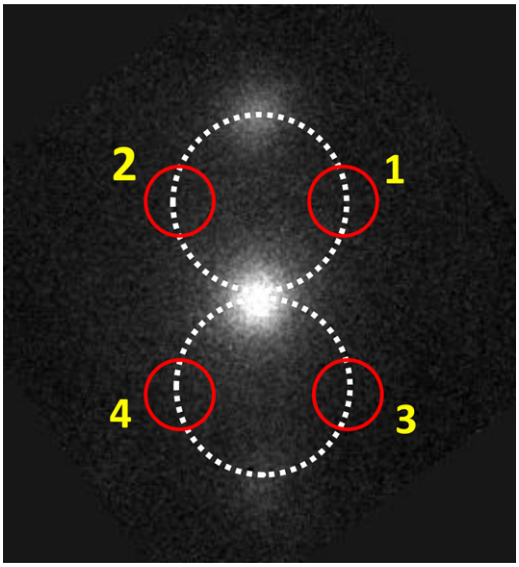

Fig. 2. ESI image showing 3-beam condition by selecting $g=200$ spot from 001 zone axis and the circles 1, 2, 3, 4 denote the positions from which the spectra can be extracted. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)